AD-A038 787 CITY COLL NEW YORK DEPT OF PHYSICS F/G 20/2
LIGHT SCATTERING IN CRYSTALS WITH SURFACE CORRECTIONS.(U)
DEC 76 M LAX, D F NELSON N00014-76-C-0099
UNCLASSIFIED TR-27 NL

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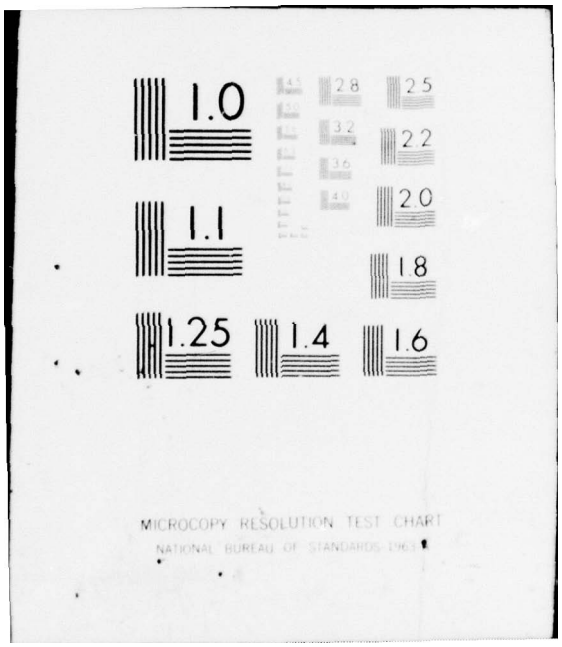
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Light Scattering in Crystals with Surface Corrections

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Office of Naval Research
Contract No. N00014-76-C-0099
Project No. NR 395-003/5-21-75
Technical Report No. 27

December 1976

unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE	READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER: 27	2. GOVT ACCESSION NO.
3. RECIPIENT'S CATALOG NUMBER: (9)	
4. TITLE (and Subtitle): (6) Light Scattering in Crystals with Surface Corrections	5. TYPE OF REPORT & PERIOD COVERED: Technical rept.
	6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s): (10) Melvin H. Lax, Donald F. Nelson	8. CONTRACT OR GRANT NUMBER(s): (15) N00014-76-C-0099
9. PERFORMING ORGANIZATION NAME AND ADDRESS: City College of CUNY, New York, N.Y. 10031	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: NR 395-003/5-21-75
11. CONTROLLING OFFICE NAME AND ADDRESS: Office of Naval Research, Physics Program Office, Arlington, Va 2217	12. REPORT DATE: (11) December 1976
	13. NUMBER OF PAGES: 13
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office): (14) TR-27 (12) 15p.	15. SECURITY CLASS. (of this report): unclassified
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

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17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

nonlinear optics
incident beam
Greens function
Raman scattering
scattered beam
Brillouin scattering
Light scattering

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

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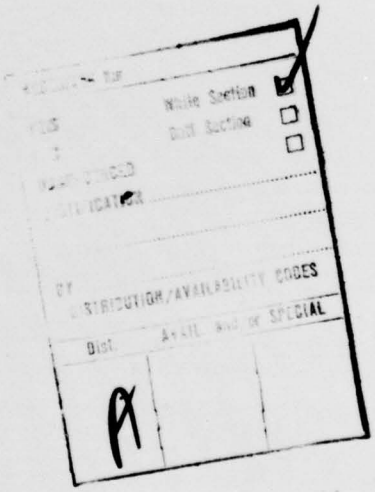

LIGHT SCATTERING IN CRYSTALS WITH SURFACE CORRECTIONS

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A summary is given of techniques and results for calculating light scattering in anisotropic crystals valid for arbitrary directions of the incident beam, the scattered beam and the crystal surface normal relative to the crystal axes. A dyadic Green's function that distinguishes ray and propagation directions leads to a scattering efficiency *inside* the crystal that involves the Gaussian curvature of the surface of wave normals. After taking account of the solid angle expansion, and the source volume demagnification at the crystal surface, a scattering formula is given suitable for comparison with experiments done *outside* the crystal. Application is made to Brillouin and Raman scattering.

I. INTRODUCTION

The availability of laser sources has led to a great upsurge in light scattering experiments in recent years. Many experiments have been performed on crystals of low symmetry and large optical anisotropy while a few experiments[1,2] have used light scattering to measure *quantitatively* the nonlinear polarization causing the scattering. Such numerical measurements require a theory which (1) accounts correctly for the noncollinearity of the ray vector and the wavevector that results from the optical anisotropy and (2) relates the nonlinear polarization to measuring instrument parameters which are outside the crystal. Previous theories[3] have often mishandled the first requirement and have always ignored the second.

The purpose of our study of the theory of light scattering, of which we present a summary here, has been to remedy these deficiencies of previous theories. To remedy the first deficiency resulting from optical anisotropy we have based our theory on a Green's function solution of the electric field wave equation.[4,5] This technique is appropriate because the scattering volume is typically very small compared to the size of the crystal under study when laser sources are used. Because even the crystal surface is in the far field, an asymptotic evaluation of the Green's function inside the crystal is adequate. Our first - and incorrect - evaluation[4] used a stationary phase integration over two variables followed by a residue integration over the third variable. The appealing but nevertheless incorrect result was what would have been calculated for an isotropic medium with the subsequent replacement of the isotropic refractive index by the one appropriate to the correct ray direction in the crystal. This replacement procedure has been used by others.[6] Our second - and correct - evaluation[5,7] follows that of Kogelnik[8] and Kogelnik and Motz[9] who used it on a magnetoionic media problem. Their work was based on a stationary phase technique of Lighthill[10] in which the residue integration was performed first and the stationary phase method was applied second. The correct asymptotically evaluated Green's function in an anisotropic crystal[5,7] is presented in Sec. II. This procedure takes proper account of the noncollinearity of the ray and propagation directions in an anisotropic crystal. In Sec. III the Green's function solution is used to find an expression for the scattered power inside the crystal. The expression differs in several ways from the best previous treatment of Brillouin scattering in anisotropic media by Motulevich.[11] One difference is the appearance of the Gaussian curvature K of the surface $\omega(\mathbf{k}) = \omega$ in our formula for the scattered power inside the crystal. Motulevich's treatment was not based on a Green's function approach but rather on Ginzburg's[12] Hamiltonian approach using an aniso-

tropic Coulomb gauge.

In Secs. IV and V we present the remedy to the second deficiency of previous theories, that is, to relating the scattered power inside the crystal to quantities characterizing the detector outside the crystal. In Sec. IV the solid angle inside the crystal is related to the expanded solid angle outside the crystal. The formulas presented are completely general in applying to any orientation of the crystal axes, the scattered ray, and the surface normal. In Sec. V the length of the scattering volume along the incident beam is related to the corresponding demagnified length as seen outside the crystal by a field stop of the detection optics. The demagnification formula is also completely general in applying to any orientation of the crystal axes, the scattered ray, and the surface normal. The width of the scattering volume (the unscattered beam) when laser sources are used is typically much less than the width of the field stop of the detection optics and so does not enter the formulas explicitly. The solid angle expansion and source volume demagnification expressions are then combined with the scattered power formula of Sec. III to produce in Sec. VII a scattered power formula applying to measurements made *outside* the crystal.

The scattered power formulas of Secs. III and VII are formulated in terms of an arbitrary mechanism of light scattering. In Sec. VIII the power formula is specialized to two important mechanisms, Brillouin scattering[1,13] (from acoustic phonons) and Raman scattering[14] (from optic phonons). The final formulas are completely general yet compact and convenient.

II. INSIDE GREEN'S FUNCTION

The wave equation for the electric field $\mathbf{E}(\mathbf{r})\exp(-i\omega t)$ in an anisotropic dielectric generated by the nonlinear polarization $\mathbf{P}^{NL}(\mathbf{r}')\exp(-i\omega t)$ at the single frequency ω is

$$\nabla\times(\nabla\times\mathbf{E}) - \frac{\omega^2}{c^2}\overleftrightarrow{\kappa}(\omega)\cdot\mathbf{E} = \frac{\omega^2}{c^2}\frac{\mathbf{P}^{NL}}{\epsilon_o}. \tag{2.1}$$

It has the Green's function solution[4]

$$\mathbf{E}(\mathbf{r},\omega) = \int \overleftrightarrow{G}(\mathbf{r}-\mathbf{r}')\cdot\mathbf{P}^{NL}(\mathbf{r}')d\mathbf{r}'e^{-i\omega t}/\epsilon_o \tag{2.2}$$

where

$$\overleftrightarrow{G}(\mathbf{R}) = \int \frac{\exp(i\mathbf{k}\cdot\mathbf{R})}{\overleftrightarrow{\alpha}(\mathbf{k},\omega)}\frac{d\mathbf{k}}{(2\pi)^3}, \tag{2.3}$$

$$\overleftrightarrow{\alpha}(\mathbf{k},\omega) \equiv (c/\omega)^2[k^2\overleftrightarrow{1} - \mathbf{kk}] - \overleftrightarrow{\kappa}(\omega), \tag{2.4}$$

and $\overleftrightarrow{\kappa}(\omega)$ is the frequency dependent dielectric tensor.

We have previously given[7] an asymptotic evaluation of Eq. (2.3) valid for $kR >> 1$. With a slight change of notation, the asymptotic dyadic Green's function can be written

$$\overleftrightarrow{G}(\mathbf{R}) = \left(\frac{\omega}{c}\right)^2 \sum_{\phi=1,2}\frac{\mathbf{e}^\phi\mathbf{e}^\phi}{\cos^2\delta^\phi}g^\phi(\mathbf{R}) \tag{2.5}$$

where $\mathbf{e}^\phi$ is the unit electric field vector associated with a given mode ϕ (e.g. extraordinary) whose ray direction $\mathbf{t}$ is parallel to the direction of observation

$$\mathbf{t} = \mathbf{R}/R, \tag{2.6}$$

and δ^ϕ is the angle between the ray vector $\mathbf{t}$ and the wavevector $\mathbf{k}^\phi$ associated with the above ray direction. The scalar Green's function becomes

$$g^{\phi}(\mathbf{R}) = f^{\phi}\frac{\exp[i\mathbf{k}^{\phi}\cdot\mathbf{R}]}{4\pi R}. \tag{2.7}$$

where

$$f^{\phi} \equiv \frac{\cos\delta^{\phi}}{k^{\phi}\sqrt{K^{\phi}}} \tag{2.8}$$

and K^{ϕ} is the Gaussian curvature of the $\omega(\mathbf{k})$ surface at $\mathbf{k}^{\phi}$. Equation (2.7) has been shown[7] to agree precisely, in the uniaxial case, with the Green's function obtained without asymptotic approximations. This Green's function disagrees by the factor f^{ϕ} with the intuitive notion that for a given direction of observation one may use the Green's function for an isotropic medium with the index of refraction appropriate to direction $\mathbf{k}^{\phi}$.

III. SCATTERED POWER INSIDE THE CRYSTAL

If Eqs. (2.2), (2.5), (2.7) and (2.8) are used to calculate the electric field and if the corresponding magnetic field, $\mathbf{H} = (\nabla\times\mathbf{E})/(i\omega\mu_o)$, is also found, then the Poynting vector for a given mode ϕ is found to be

$$\mathbf{S}^{\phi} = \frac{\mu_o\omega^4}{32\pi^2 c}\frac{n^{\phi}|f^{\phi}|^2|c^{\phi}|^2\mathbf{t}}{(\cos\delta^{\phi})^3} \tag{3.1}$$

where n^{ϕ} is the index of refraction appropriate to wavevector $\mathbf{k}^{\phi}$ and

$$c^{\phi} \equiv \mathbf{e}^{\phi}\cdot\int_{VS}\mathbf{P}^{NL}(\mathbf{r})\exp(-i\mathbf{k}^{\phi}\cdot\mathbf{r})\,d\mathbf{r}. \tag{3.2}$$

The ratio of the scattered power inside the crystal,

$$P_{\text{ins}}^{\text{scat}} = |\mathbf{S}^{\phi}|r^2 d\Omega^{r}, \tag{3.3}$$

(where $d\Omega^{r}$ is a solid angle of rays in $\mathbf{r}$ space) to the incident power,

$$P_{\text{ins}}^{\text{inc}} = A|\mathbf{S}^{u}| = A\frac{1}{2}cn^{u}|\mathbf{E}^{u}|^2\cos\delta^{u}, \tag{3.4}$$

may be expressed as

$$\frac{P_{\text{ins}}^{\text{scat}}}{P_{\text{ins}}^{\text{inc}}/A} = RV^{S}d\Omega_{\text{in}}^{r}. \tag{3.5}$$

Here V^S is the scattering volume accepted by the detector, A, the cross-sectional area of the incident beam inside the crystal and R, the scattering efficiency (the scattered power per unit incident power, per unit solid angle, per unit path length), is given by

$$R = \left(\frac{\omega}{c}\right)^4\frac{n^{\phi}}{8\pi^2 n^{u}\cos\delta^{\phi}\cos\delta^{u}}\frac{J}{(k^{\phi})^2K^{\phi}}. \tag{3.6}$$

The nonlinear phenomena that give rise to the scattering are included in the quantity

$$J = \frac{\left|\mathbf{e}^{\phi}\cdot\int_{VS}\mathbf{P}^{NL}(\mathbf{r})\,e^{-i\mathbf{k}^{\phi}\cdot\mathbf{r}}d\mathbf{r}\right|^2}{2\epsilon_o^2|\mathbf{E}^{u}|^2V^{S}}. \tag{3.7}$$

One striking way that the expression, Eq. (3.6), for the scattering efficiency differs from previous expressions is by its dependence on the Gaussian curvature K^{ϕ} of the ϕ branch of the $\omega(\mathbf{k}) = \omega$ surface.

IV. SOLID ANGLE EXPANSION

The ratio of the solid angles subtended by the rays of a beam inside and outside a crystal can be factored as

$$\frac{d\Omega^r_{\text{in}}}{d\Omega_{\text{out}}} = \frac{d\Omega^r_{\text{in}}}{d\Omega^k_{\text{in}}} \frac{d\Omega^k_{\text{in}}}{d\Omega_{\text{out}}} . \tag{4.1}$$

The first factor describes the ratio of the solid angles in ray vector space and wavevector space and is independent of the existence of a crystal surface. The second factor, describing the change in wavevector solid angles, is completely determined by Snell's law.

Since $d\Omega^k_{\text{in}}$ is readily computed in terms of the area of a patch dA^k of the $\omega(\mathbf{k})$ surface, see Fig. 2, and $d\Omega^r_{\text{in}}$ is related to the same area by Gauss' *theorema egregium*, see Fig. 3, we obtain our previously quoted result[7]

$$d\Omega^r_{\text{in}}/d\Omega^k_{\text{in}} = K^\phi (k^\phi)^2/\cos\delta^\phi . \tag{4.2}$$

Across the surface of a crystal (nominally in the 3 or z direction) Snell's law guarantees the continuity of the transverse components of the propagation vector and, hence, of $dk_1\, dk_2$. Since $dk_1\, dk_2$ is simply related to the patch area dA^k, see Fig. 4, and hence to the solid angle $d\Omega^k$, see Fig. 2, Snell's law leads to the relation[7]

$$\frac{d\Omega^k_{\text{in}}}{d\Omega_{\text{out}}} = \frac{\cos\delta^\phi \cos\alpha}{(n^\phi)^2 \cos\beta} , \tag{4.3}$$

where β is the angle of arrival of the ray inside the crystal to the surface normal, δ^ϕ is the angle between ray and wavevectors, as before, and α is the angle between the departure ray outside the crystal and the surface normal.

The product of Eqs. (4.2) and (4.3) yields the desired solid angle expansion

$$d\Omega^r_{\text{in}}/d\Omega_{\text{out}} = (\omega/c)^2 K^\phi \cos\alpha/\cos\beta . \tag{4.4}$$

A slight rearrangement of this equation suggests that

$$d\Omega' \cos\beta/K \tag{4.5}$$

is an invariant for the passage of a beam from one material to another, a result we have recently proved quite generally.[15] Equation (4.4) is a special case of this invariance in which the second medium is a vacuum with $K_{\text{vac}} = (c/\omega)^2$.

V. SOURCE VOLUME DEMAGNIFICATION

When the laser beam, scattered ray, surface normal, and departure ray outside the crystal are all in one plane, it is possible to derive the length l_S along the laser beam in the crystal from which radiation is admitted by a detector field stop of length l_D. As seen from Fig. 5, these lengths are related by

$$\frac{l_S \sin\theta_S}{\cos\beta} = \frac{l}{\cos\beta} = \frac{l_D}{\cos\alpha} \tag{5.1}$$

where θ_S is the scattering angle.

When the rays mentioned above are not all coplanar, Eq. (5.1) must be replaced by

$$\frac{l_S}{l_D} = \frac{N\cos\beta}{\sin\theta_S \cos\alpha} \tag{5.2}$$

where N is the noncoplanarity correction,[16]

$$N = 1/(\cos\phi \cos\phi' - \cos\beta \sin\phi \sin\phi') . \tag{5.3}$$

Here ϕ' is the angle of tilt between the plane defined by the unscattered laser beam and the normal to the input surface and the plane defined by the scattered ray and exit surface normal; ϕ is the angle of tilt between the latter plane and the departure plane defined by the normal to the exit surface and the scattered ray after it has left the crystal.

VI. SCATTERED POWER OUTSIDE THE CRYSTAL

To convert Eq. (3.5) to a power scattering formula outside the crystal, we use

$$P_{\text{ins}}^{\text{scat}} = P_{\text{out}}^{\text{scat}} / T^{\text{exit}} , \tag{6.1}$$

$$P_{\text{ins}}^{\text{inc}} = P_{\text{out}}^{\text{inc}} T^{\text{ent}} \tag{6.2}$$

where T^{ent} and T^{exit} are entrance and exit transmission factors. If we note that $V^S/A = l_S$, Eqs. (4.4) and (5.2) can be combined to yield

$$l_S d\Omega_{\text{in}}^{r} = \left(\frac{\omega}{c}\right)^2 \frac{N l_D d\Omega^D K^{\phi}}{\sin\theta_S} \tag{6.3}$$

where we have written $d\Omega^D$ for $d\Omega_{\text{out}}$ to remind us that it is the detector solid angle. A detailed derivation of this result has been given elsewhere.[7,14,15,16]

If Eqs. (6.1), (6.2) and (6.3) are combined with Eq. (3.5), we obtain

$$\frac{P_{\text{out}}^{\text{scat}}}{P_{\text{out}}^{\text{inc}}} = \frac{R T^{\text{ent}} T^{\text{exit}} l_D d\Omega^D N K^{\phi}}{\sin\theta_S} \left(\frac{\omega}{c}\right)^2 . \tag{6.4}$$

If Eq. (3.6) is used for the scattering efficiency, we obtain

$$\frac{P_{\text{out}}^{\text{scat}}}{P_{\text{out}}^{\text{inc}}} = \left(\frac{\omega}{c}\right)^4 \frac{N l_D d\Omega^D}{\sin\theta_S} \frac{T^{\text{ent}} T^{\text{exit}} J}{8\pi^2 n^{\phi} n^{\theta} \cos\delta^{\phi} \cos\delta^{\theta}} . \tag{6.5}$$

Equation (6.5) incorporates all the geometric optics of the crystal surface. The mechanism of the scattering is contained in J, whose evaluation will be discussed in the next section.

VII. APPLICATION TO LIGHT SCATTERING

To apply our final scattering equation, Eq. (6.5), to Brillouin, Raman, or some other scattering mechanism, it is necessary to evaluate J, of Eq. (3.7), which involves the nonlinear optical properties of the scattering medium. Since the volume V^S is large compared to all relevant wavelengths, it is permissible to take the limit as V^S approaches infinity. The Weiner-Khinchin theorem[17] applied to spatial variables rather than the time then permits Eq. (3.7) to be rewritten as an autocorrelation,

$$J = \frac{e_i^{\phi} e_a^{\phi} \int \exp(-i\mathbf{k}^{\phi} \cdot \mathbf{r}) < P_i^{NL}(0)^* P_a^{NL}(\mathbf{r}) >}{2\epsilon_o^2 |\mathbf{E}^{\theta}(\mathbf{r})|^2} , \tag{7.1}$$

where the limits are infinite.

With the understanding

$$\mathbf{E}^{\theta}(\mathbf{r},t) = \frac{1}{2}\left[\mathbf{E}^{\theta}(\mathbf{r}) e^{-i\omega_L t} + \mathbf{E}^{\theta}(\mathbf{r})^* e^{i\omega_L t}\right] , \tag{7.2}$$

$$\mathbf{P}^{NL}(\mathbf{r},t) = \frac{1}{2}\left[\mathbf{P}^{NL}(\mathbf{r}) e^{-i\omega_B t} + \mathbf{P}^{NL}(\mathbf{r})^* e^{i\omega_B t}\right] , \tag{7.3}$$

where ω_L is the input or laser frequency and ω_B is the Brillouin (or Raman) scattered frequency, we can write

$$P_a^{NL}(\mathbf{r},t) = \epsilon_o \chi_{ab}(\mathbf{r},t) E_b^{\theta}(\mathbf{r},t) \tag{7.4}$$

where the "susceptibility" χ_{ab} is space and time dependent because it is induced by the phonon field in Brillouin or Raman scattering. Equation (7.1) can then be simplified to

$$J = \frac{1}{2}\int \exp\left[-i(\mathbf{k}^{\phi}-\mathbf{k}^{\theta})\cdot\mathbf{r}\right] < N^{\phi\theta}(0,0)\, N^{\phi\theta}(\mathbf{r},0) > d\mathbf{r} \tag{7.5}$$

where

$$N^{\phi\theta}(\mathbf{r},t) = e_a^{\phi}\chi_{ab}(\mathbf{r},t) e_b^{\theta} \tag{7.6}$$

and we have used the plane-wave character $\exp(i\mathbf{k}^{\theta}\cdot\mathbf{r})$ of the unscattered wave $\mathbf{E}^{\theta}(\mathbf{r})$.

If one is concerned with line shape, Eq. (7.5) can be decomposed by

$$J = \int_0^{\infty} J(\omega)\, d\omega/2\pi \tag{7.7}$$

where

$$J(\omega) = \frac{1}{2}\int\int \exp\left[i(\omega-\omega_L)t\right] \exp\left[-i(\mathbf{k}^{\phi}-\mathbf{k}^{\theta})\cdot\mathbf{r}\right] < N^{\phi\theta}(0,0)\, N^{\phi\theta}(\mathbf{r},t) > d\mathbf{r}\, dt, \tag{7.8}$$

and only the positive frequency components of $\exp(-i\omega_L t)\, N^{\phi\theta}(\mathbf{r},t)$ are included so that $J(\omega)$ vanishes for $\omega < 0$.

Brillouin Scattering

The first application of our modified Green's function, Eq. (2.7), with surface corrections was made in connection with a detailed study of Brillouin scattering in calcite. Because the formulas in this paper give a clear factorization of our Brillouin scattering formula[1] into intrinsic and geometric components, we indicate here the evaluation of J based on Eq. (7.5). To be consistent with our definition[1] of the photoelastic susceptibility as a relation between the positive frequency components of $\mathbf{P}^{NL}$, $\mathbf{E}^{\theta}$ and $u_{c,d}$ (the displacement gradient), we must write

$$\chi_{ab} = 2\chi_{abcd}\, u_{c,d}\,. \tag{7.9}$$

The averages, $< u_{i,j}(0,0)\, u_{k,l}(\mathbf{r},t) >$, can be evaluated by using the expansion of u_c in terms of normal coordinates,[18]

$$\mathbf{u}(\mathbf{r},t) = \sum_{\mathbf{q}}\left[\frac{\hbar}{2\rho\Omega\,\omega(\mathbf{q})}\right]^{\frac{1}{2}} \mathbf{b}\left[e^{i\mathbf{q}\cdot\mathbf{r}}\, a(\mathbf{q},t) + e^{-i\mathbf{q}\cdot\mathbf{r}}\, a^{\dagger}(\mathbf{q},t)\right], \tag{7.10}$$

where $\mathbf{q} \equiv \mathbf{k}^{A}$ is the acoustic phonon propagation vector, ρ is the crystal density and Ω is its volume so that $\rho\Omega = MN$ = mass per unit cell × number of cells. The unit displacement vector $\mathbf{b}$ is characteristic of the type of mode (e.g. transverse), and the sum over $\mathbf{q}$ also implies a sum over types of modes. The mode amplitudes in the quantum mechanical case obey

$$< a^{\dagger}(\mathbf{q},t)\, a(\mathbf{q}',t) > = \bar{n}\,\delta(\mathbf{q},\mathbf{q}')\,, \tag{7.11}$$

$$< a(\mathbf{q},t)\, a^{\dagger}(\mathbf{q}',t) > = (\bar{n}+1)\,\delta(\mathbf{q},\mathbf{q}') \tag{7.12}$$

where $\bar{n}$ is the actual phonon excitation number that reduces to

$$\bar{n} = \frac{1}{\exp\left[\hbar\omega(\mathbf{q})/kT\right]-1} \tag{7.13}$$

in the thermal equilibrium case. Averages of the type, Eq. (7.12), contribute to Stokes scattering, whereas those of type, Eq. (7.11), contribute to anti-Stokes scattering. Because we usually have $\hbar\omega << kT$, both Stokes and anti-Stokes scattering have an intensity proportional $\bar{n} \approx kT/\hbar\omega$. If we combine Eqs. (7.5) and (7.9)-(7.13), we obtain

$$J = \frac{kT}{\rho v_A^2} G, \quad G \equiv | \, e_i^\phi \, e_j^\theta \, \chi_{ijkl} \, b_k \, a_l \, |^2 \tag{7.14}$$

where $a_l \equiv q_l / |\mathbf{q}| = k_l^A / |\mathbf{k}^A|$ is a unit vector in the direction of phonon propagation and

$$v_A \equiv \omega(\mathbf{k}^A)/k^A \tag{7.15}$$

is the sound velocity.

Equation (6.5) for the Stokes (or anti-Stokes) power scattering by a single type of acoustic mode can be rewritten as

$$\frac{P_{\text{out}}^{\text{scat}}}{P_{\text{out}}^{\text{inc}}} = \left(\frac{\omega}{c}\right)^4 \frac{N l_D \, d\Omega^D}{\sin\theta_S} \frac{T^{\text{ent}} T^{\text{exit}} \, kTG}{8\pi^2 \, n^\phi \, n^\theta \cos\delta^\phi \cos\delta^\theta \rho v_A^2}, \tag{7.16}$$

where G of Eq. (7.14) can also be expressed in terms of the Pockels tensor, p_{ijkl}, by means of

$$G = (1/4) \, (n^\phi \, n^\theta)^4 \, (\cos\delta^\phi \cos\delta^\theta)^2 \, F, \tag{7.17}$$

$$F \equiv | \, d_i^\phi \, d_j^\theta \, p_{ijkl} \, b_k \, a_l \, |^2 . \tag{7.18}$$

Here $\mathbf{d}^\theta$ and $\mathbf{d}^\phi$ are unit electric displacement vectors of the input and scattered beams, and ω is the Brillouin scattered frequency ω^B. Equation (7.16) was used in the analysis of our experimental results[1,19] except for the noncoplanarity factor, N, which was not needed then since all experiments were conducted in a symmetry plane.

Raman Scattering

A detailed analysis of Raman scattering by polaritons using the fluctuation-dissipation approach of Barker and Loudon[6] has been given.[14] To evaluate Eq. (7.8) for $J(\omega)$, needed to obtain the line shape in Raman scattering, we express the nonlinear susceptibility defined in Eq. (7.4) in the usual way,

$$\chi_{ij}(\mathbf{r},t) = 2\sum_\mu A_{ij}^\mu \, w^\mu(\mathbf{r},t) + 2B_{ijk} \, E_k(\mathbf{r},t), \tag{7.19}$$

as an ionic contribution associated with the displacement w^μ of mode μ plus an electronic contribution proportional to the electric field $E(\mathbf{r},t)$. The factors of 2 assure conformity with definitions in our previous work.[14] Because field and displacement are correlated in polariton motion, it was simpler to evaluate the correlations with the help of the fluctuation-dissipation theorem.[14,6] If T_{BA} represents the response of $\langle B \rangle$ to a unit force at frequency ω applied to A, Eq. (7.8) yields[20]

$$J(\omega + \omega^L) = 4\hbar \, n(\omega) \, e_i^\phi \, e_j^\theta \, e_a^\phi \, e_b^\theta \, \text{Im}[\, J_{ijab} \,] \tag{7.20}$$

where

$$J_{ijab} = \sum_{\mu\nu} (A_{ij}^\mu)^* A_{ab}^\nu \, T_{\nu\mu'} + B_{ijk}^* \, B_{abc} \, T_{ck'} + \sum_\mu (A_{ij}^\mu)^* B_{abc} \, T_{c\mu'} + \sum_\nu B_{ijk}^* \, A_{ab}^\nu \, T_{\nu k'} \tag{7.21}$$

$$= \left[\sum_\mu (A_{ij}^\mu)^* \, q_k^\mu \, B^\mu(\omega) + B_{ijk}^*\right] T_{ck'} \left[\sum_\nu A_{ab}^\nu \, q_c^\nu \, B^\nu(\omega) + B_{abc}\right]$$

$$+ \sum_\mu (A_{ij}^\mu)^* A_{ab}^\mu \, B^\mu(\omega) + \sum_\mu (A_{ij}^\mu)^* B^\mu(\omega) . \tag{7.22}$$

The second form, Eq. (7.22), has used the equations of motion to express all response functions in terms of the field-field response, i.e. the response of the electric field to a unit applied external polarization, as

$$T_{ck'} = E_c / P_k^{ext} = (\overleftrightarrow{\alpha}^{-1})_{ck} / \epsilon_o$$

$$= \sum_{\phi=1,2} \frac{e_c^\phi \; e_k^\phi}{\epsilon_o \cos^2 \delta^\phi \; [(ck/\omega)^2 - (n^\phi)^2]} - \frac{s_c \; s_k}{\epsilon_o \; \mathbf{s} \cdot \overleftrightarrow{\kappa}(\omega) \cdot \mathbf{s}} \tag{7.23}$$

The first two terms in Eq. (7.23) represent the transverse modes while the third term represents the longitudinal mode.

The mode w^μ will generally obey an equation of the form[20]

$$m^\mu \; [(\omega^\mu)^2 - \omega^2 - i\omega \; \gamma^\mu(\omega)] \; w^\mu = q_c^\mu \; E_c \tag{7.24}$$

where m^μ is the effective mass, ω^μ the effective (angular) frequency, $\gamma^\mu(\omega)$ the effective frequency dependent damping constant and q_c^μ is the c component of the effective charge. The response of w^μ to a unit applied force, needed in Eq. (7.22), is given by

$$B^\mu(\omega) = \left\{ m^\mu \; [(\omega^\mu)^2 - \omega^2 - i\omega \; \gamma^\mu(\omega)] \right\}^{-1} . \tag{7.25}$$

We may combine Eqs. (7.20) – (7.25) to obtain $J(\omega + \omega^L)$. If $J(\omega + \omega^L)$ is inserted in place of J in Eq. (6.5), the result, in view of Eq. (7.7), is the ratio of the Raman scattered power per unit frequency interval to the incident power, both computed outside the crystal.

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* Supported in part by AROD and ONR.

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FIGURE CAPTIONS

FIG. 1. A typical experimental setup for a Raman scattering experiment which displays the expansion of the solid angle on emerging from the crystal. For a narrow laser beam, the source volume $V^S = Al_S$ from which scattered light is accepted is limited by the input beam area A and a length l_S determined by the field stop.

FIG. 2. The solid angle $d\Omega^k$ associated with a patch of area $dA^k = dudv$ on the surface $\omega(\mathbf{k}) = \omega$ of free-wave $\mathbf{k}$ vectors is given by $d\Omega^k = dA^k \cos\delta/k^2$, where $dA^k \cos\delta$ is the component of the area dA^k normal to $\mathbf{k}$, since dA^k is normal to $\mathbf{t}$ or $\nabla\omega(\mathbf{k})$.

FIG. 3. Gauss's bump theorem relates the area $dA = dudv$ of an element of surface to the area $d\Omega^r$ on the surface of a unit sphere subtended by the unit normals to $dudv$ that have been shifted in a parallel manner until their origins coincide at O, the sphere center. The solid angle $d\Omega^r = d\theta\, d\phi$ where $d\theta = du/\rho_u$ is the angle AUD and $d\phi = dv/\rho_v$ is the angle AVB where ρ_u and ρ_v are the principal radii of curvature. Thus $d\Omega^r = K\, dA^k$ where $K = (\rho_u\, \rho_v)^{-1}$ is the Gaussian curvature and $dA^k = dudv$ is the area of the patch on the $\omega(\mathbf{k})$ surface.

FIG. 4. The patch $dA^k = dudv$ on the surface $\omega(\mathbf{k}) = \omega$ of Fig. 3 is plotted in a k_1, k_2, k_3 coordinate system in which the three-direction is along the direction $\mathbf{n}$ of a unit normal to the surface of the crystal. The projection of dA^k onto the 1-2 plane yields $dk_1\, dk_2 = dA^k \cos\beta$ because β is the angle between the normal $\mathbf{t}$ to the patch (the ray direction) and the normal $\mathbf{n}$ to the surface.

FIG. 5. Demagnification corrections when the arrival ray, departure ray, and surface normal are all in one plane. The detector field stop (see Fig. 1) is represented in image space by the knife edges, which accept a dimension l_D perpendicular to the beam. The portion l_S of the laser beam accepted is determined by the geometrical conditions shown, independent of the orientation of the virtual image $l_A : l_S \sin\theta_S / \cos\beta = l/\cos\beta = l_D/\cos\alpha$.

This manuscript was produced on a computer driven phototypesetter using the UNIX operating system developed at Bell Laboratories.

M. Lax

MH-1154 ML
1153 DFN -ce

D. F. Nelson

Att.
Figures 1-5

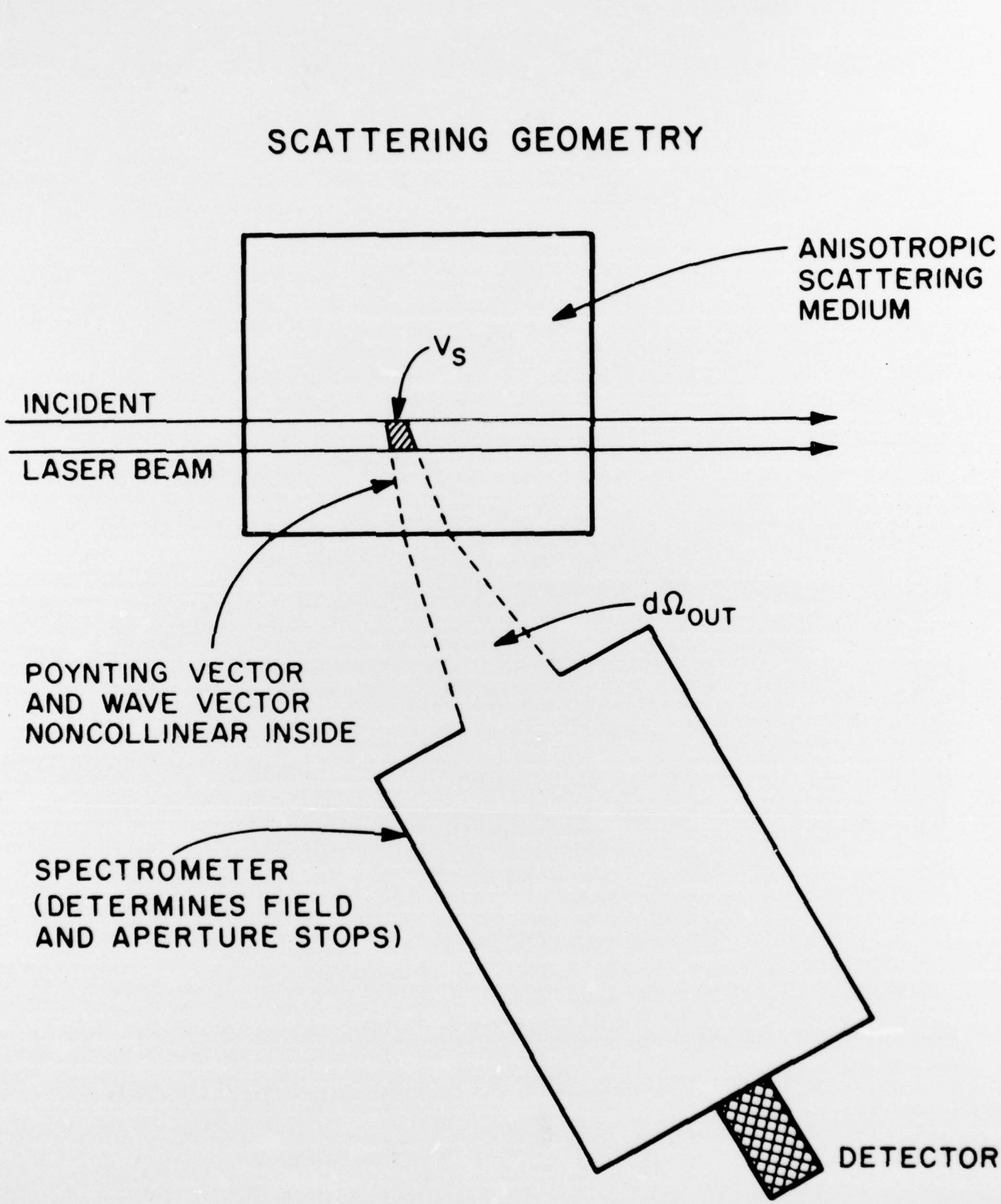

SCATTERING GEOMETRY
ANISOTROPIC SCATTERING MEDIUM
V_S
INCIDENT LASER BEAM
$d\Omega_{OUT}$
POYNTING VECTOR AND WAVE VECTOR NONCOLLINEAR INSIDE
SPECTROMETER (DETERMINES FIELD AND APERTURE STOPS)
DETECTOR

FIG. 1

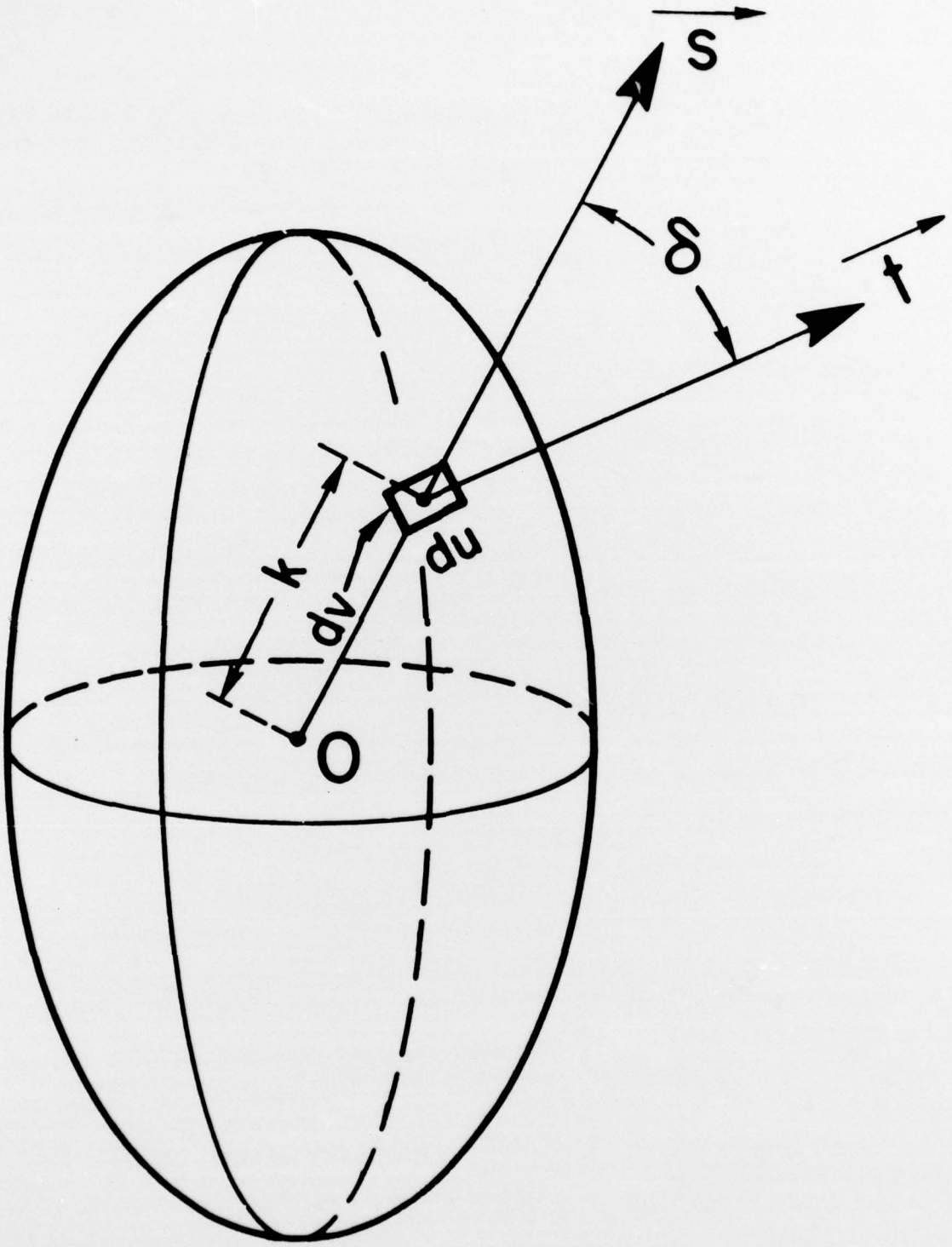
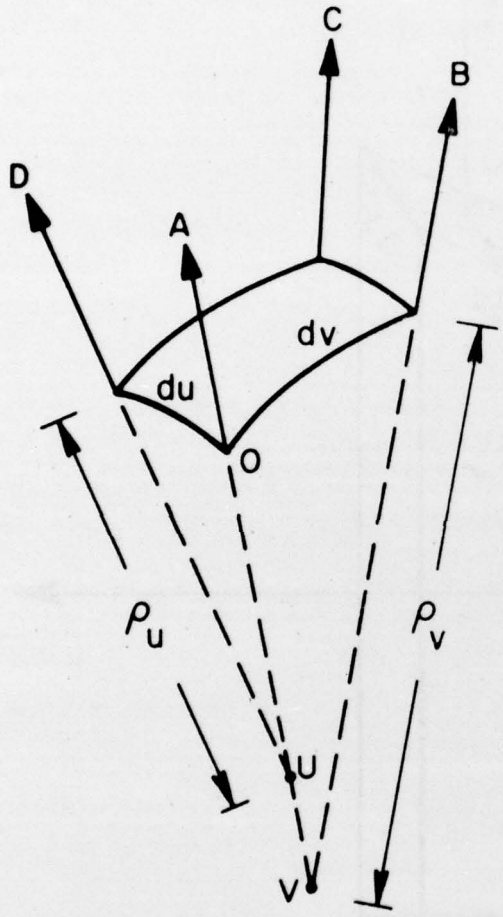
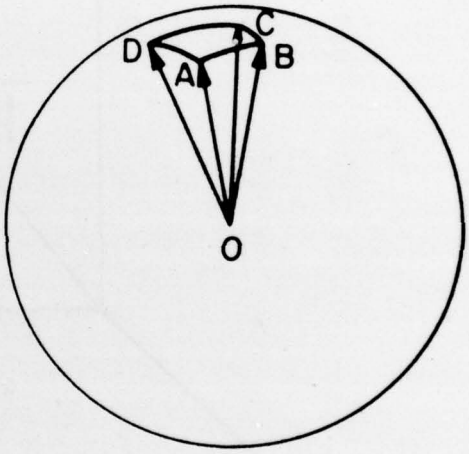


FIG. 2

GAUSS' BUMP THEOREM





$$d\Omega_r = \frac{du}{\rho_u}\frac{dv}{\rho_v} = K\,dA$$

FIG. 3

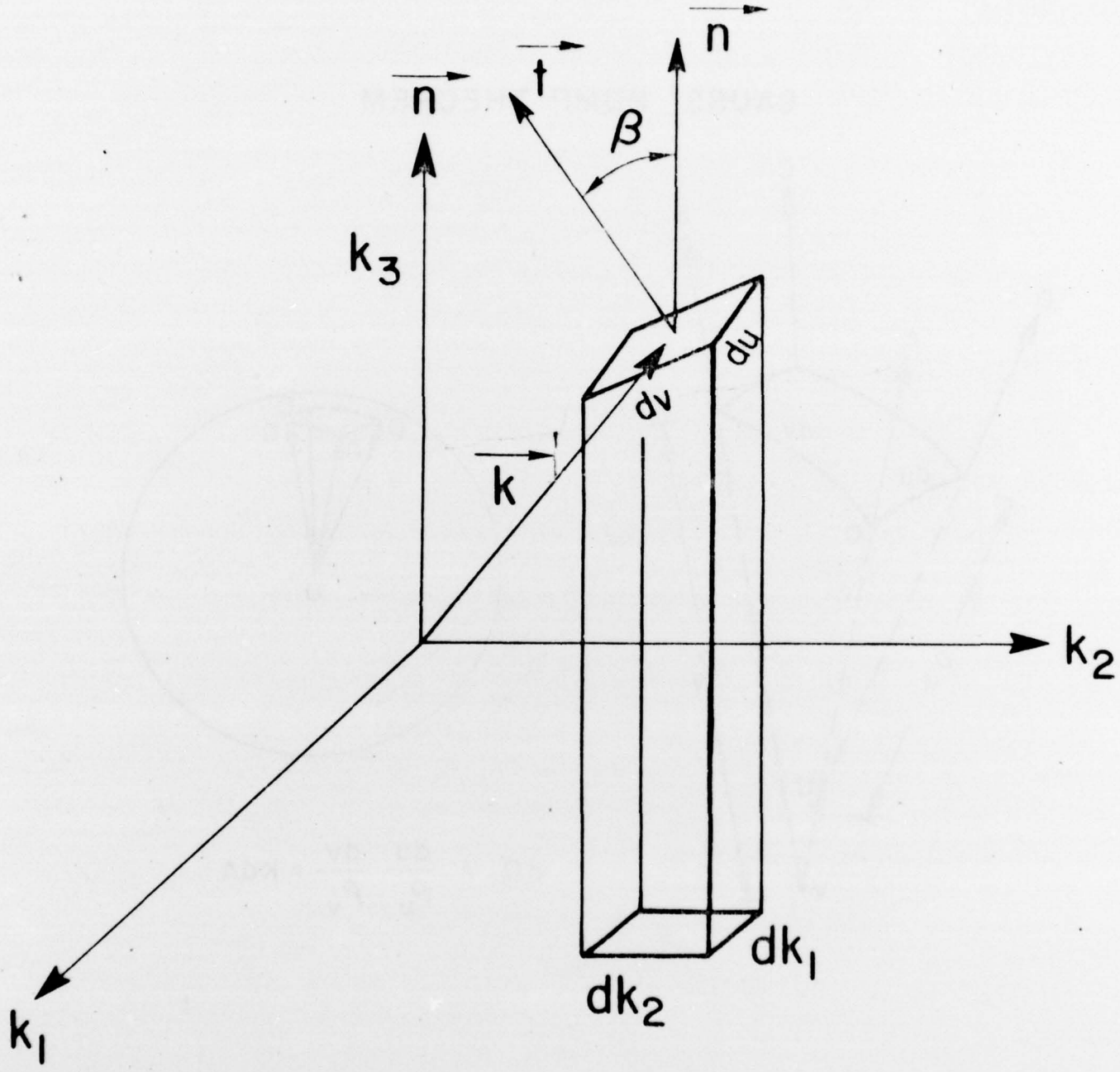
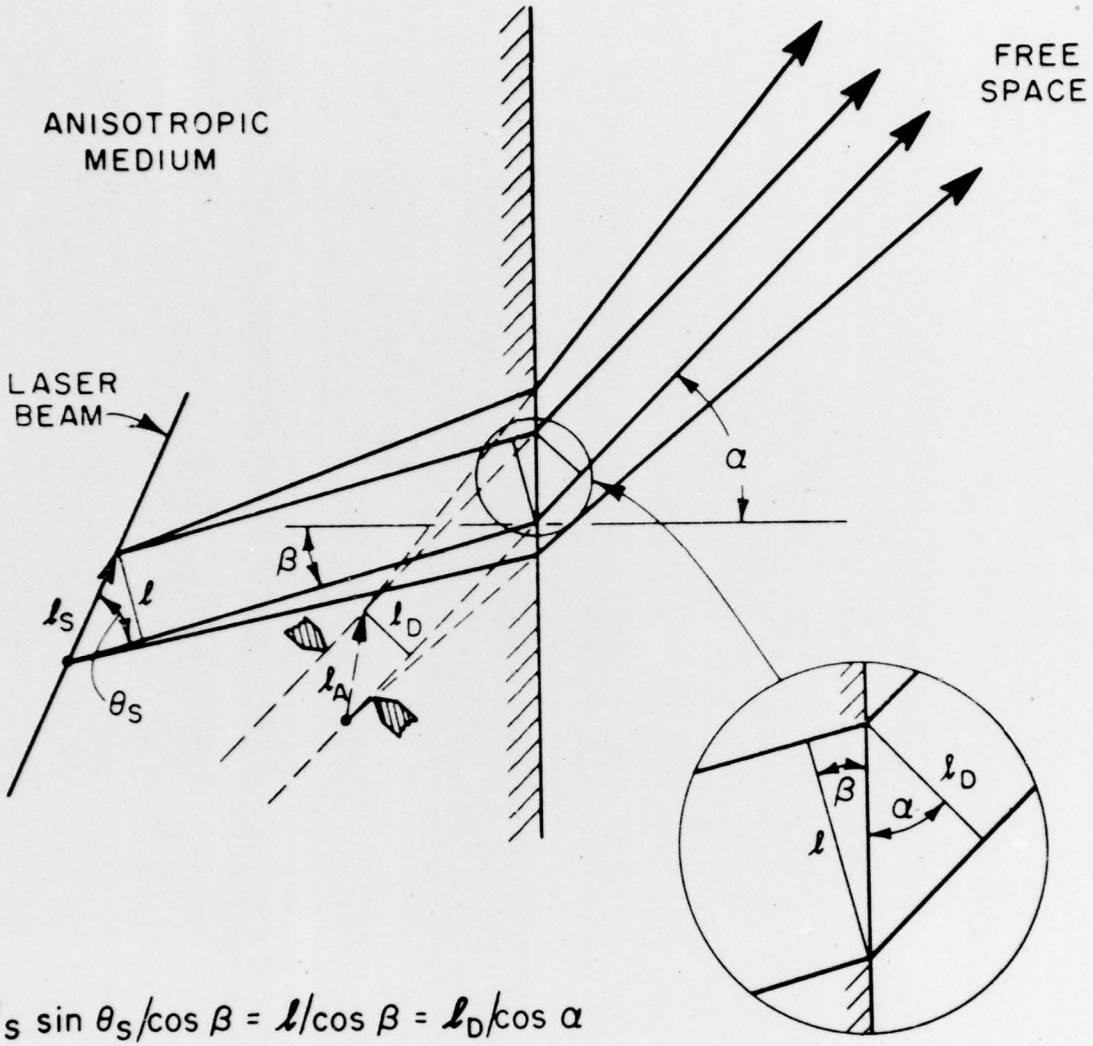


FIG. 4



$$\therefore \ \ell_S \sin\theta_S/\cos\beta = \ell/\cos\beta = \ell_D/\cos\alpha$$

FIG. 5